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(72) Inventor: **The designation of the inventor has not
yet been filed**

(74) Representative: **VA TECH Patente GmbH & Co
Stahlstrasse 21 a
4020 Linz (AT)**

(71) Applicant: **Borealis Technology OY
06201 Porvoo (FI)**

(54) **Propylene polymers with improved properties**

(57) The invention relates to novel propylene polymers with improved properties especially with improved stiffness and impact strength comprising propylene homopolymers or propylene block copolymers with 90.0 to 99.9 wt% propylene and 0.1 to 10 wt% α -olefins with 2 or 4 to 18 carbon atoms, or mixtures thereof, wherein the propylene homopolymers or propylene block copolymers are β -nucleated propylene polymers, whereby the β -nucleated propylene homopolymers have an $IR_t > 0.98$, a tensile modulus > 1500 MPa at $+23^\circ\text{C}$ and a Charpy impact strength, ≥ 3 kJ/m² at -20°C using

notched test specimens, and the β -nucleated propylene block copolymers are polymers having an IR_t of the propylene homopolymer block of > 0.98 , a tensile modulus ≥ 1100 MPa at $+23^\circ\text{C}$ and a Charpy impact strength of > 6 kJ/m², at -20°C using notched test specimens.

The propylene polymers with an improved property spectrum are suitable for producing molded parts in a pipe system, such as pipes and fittings, inspection chambers, pipe ducting systems, extrusion or compression molded sheets and the like.

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Description

Field of the invention

5 **[0001]** The invention relates to propylene polymers with improved properties as well as to a process for producing them.

Background of the invention

10 **[0002]** Propylene polymer compositions from common propylene polymers and β -nucleating agents are known. β -Nucleating agents are useful in compositions for forming unstretched films which can be made porous by extraction of β -spherulites and stretching the films (disclosed for example in US-A 4 386 129; US-A 4,975,469).

[0003] Another effect of β -nucleating agents in propylene compositions is the improved thermoformability. As the melting point of the β -form of the spherulites of polypropylene based resins is generally about 144 to 148°C, contrasted with the typical melting point range of α -form spherulites of about 159 to 163°C, melt forming from these compositions is possible at lower temperatures and higher production rates (WO 93/12 262).

[0004] The disadvantage of these compositions having good impact properties is that they show unsatisfactory strength.

Conventional polypropylene homopolymers usually show high stiffness, but poor impact strength. Conventional β -nucleated polypropylenes usually have improved impact strength but also exhibit reduced stiffness.

Object of the invention

25 **[0005]** Therefore it is the object of the present invention to provide propylene polymers with improved properties characterized in having high stiffness and good impact properties simultaneously.

Brief description of the invention

30 **[0006]** According to the present invention, this object is achieved by propylene polymers with improved properties comprising propylene homopolymers with melt indices of 0.05 to 15 g/10 min at 230°C/2.16 kg or propylene block copolymers with 90.0 to 99.9 wt% propylene and 0.1 to 10 wt% α -olefins with 2 or 4 to 18 carbon atoms with melt indices of 0.05 to 20 g/10 min at 230°C/2.16 kg, or mixtures thereof, wherein the propylene homopolymers or propylene block copolymers are β -nucleated propylene polymers, whereby the β -nucleated propylene homopolymers have an $IR_{\tau} > 0.98$, a tensile modulus of ≥ 1500 MPa at +23°C and a Charpy impact strength of ≥ 3 kJ/m² at -20°C using notched test specimens, and the β -nucleated propylene block copolymers are polymers having an IR_{τ} of the propylene homopolymer block of > 0.98 , a tensile modulus of ≥ 1100 MPa at +23 °C and a Charpy impact strength, of ≥ 6 kJ/m² at -20°C, using notched test specimens.

Detailed description of the invention

40 **[0007]** β -nucleated propylene polymers are isotactic propylene polymers composed of chains in a 3_1 helical conformation having an internal microstructure of β -form spherulites being composed of radial arrays of parallel stacked lamellae. This microstructure can be realized by the addition of β -nucleating agents to the melt and subsequent crystallization. The presence of the β -form can be detected through the use of wide angle X-ray diffraction (Moore, J., Polypropylene Handbook, p. 134-135, Hanser Publishers Munich 1996).

45 **[0008]** The IR_{τ} of the propylene polymers is determined by Infrared spectroscopy and calculated as described in EP 0 277 514 A2 on page 3.

[0009] The propylene homopolymers according to the present invention have melt indices of 0.05 to 15 g/10 min at 230°C/2.16 kg, preferably 0.1 to 8 g/10 min at 230°C/2.16 kg, most preferably 0.2 to 5 g/10 min at 230°C/2.16 kg.

50 The propylene copolymers according to the present invention have melt indices of 0.05 to 20 g/10 min at 230°C/2.16 kg, preferably 0.1 to 8 g/10 min at 230°C/2.16 kg, most preferably 0.2 to 5 g/10 min at 230°C/2.16 kg.

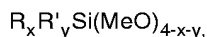
[0010] According to the present invention the propylene homopolymers show a tensile modulus ≥ 1500 MPa, preferably ≥ 1600 MPa and the propylene copolymers show a tensile modulus ≥ 1100 MPa, preferably ≥ 1300 MPa and most preferably ≥ 1500 MPa.

55 **[0011]** The propylene homopolymers according to the present invention have a Charpy impact strength of ≥ 3 kJ/m² at -20°C, preferably 4 to 10 kJ/m² at -20°C, most preferably 5 to 10 kJ/m² at -20°C.

The propylene copolymers according to the present invention have a Charpy impact strength of ≥ 6 kJ/m² at -20°C, preferably ≥ 9 kJ/m² at -20°C, most preferably ≥ 10 kJ/m² at -20°C. Charpy impact strength of up to at least 60 kJ/m² is

possible for copolymers according to the invention.

[0012] According to an advantageous feature of the present invention the β -nucleated propylene polymers with an $IR\tau > 0.98$ being propylene polymers obtained by polymerization with a Ziegler-Natta catalyst system comprising titanium-containing solid components, an organoalumina, magnesium or titanium compound as cocatalyst and an external donor according to the formula



wherein R and R' are identical or different and are branched or cyclic aliphatic or aromatic hydrocarbon residues, and y and x independently from each other are 0 or 1, provided that x + y are 1 or 2.

[0013] Examples of propylene polymers with high $IR\tau$ obtained by polymerization with a Ziegler-Natta catalyst system, for example by slurry, bulk or gas phase polymerization, are propylene polymers as described in EP-A-0,790,262, WO 99/24,478 and WO 99/16,797.

[0014] A preferred external donor of the Ziegler-Natta catalyst system is dicyclopentyl dimethoxysilane.

[0015] According to a preferred embodiment the β -nucleated propylene polymer contains 0.01 to 2.0 wt%, in each case based on the propylene polymers used, of

- dicarboxylic acid derivative type diamide compounds from C_5 - C_8 -cycloalkyl monoamines or C_6 - C_{12} -aromatic monoamines and C_5 - C_8 -aliphatic, C_5 - C_8 -cycloaliphatic or C_6 - C_{12} -aromatic dicarboxylic acids, preferably N,N'-di- C_5 - C_8 -cycloalkyl-2,6-naphthalene dicarboxamide compounds, N,N'-di- C_5 - C_8 -cycloalkyl-4,4-biphenyldicarboxamide compounds, N,N'-di- C_5 - C_8 -cycloalkylterephthalamide compounds, N,N'-di- C_5 - C_8 -cycloalkyl-1,4-cyclohexanedicarboxamide compounds and/or N,N'-di- C_6 - C_{12} -aryl- C_5 - C_8 -diamide compounds; and/or
- diamine derivative type diamide compounds from C_5 - C_8 -cycloalkyl monocarboxylic acids or C_6 - C_{12} -aromatic monocarboxylic acids and C_5 - C_8 -cycloaliphatic or C_6 - C_{12} -aromatic diamines, preferably N,N'- C_6 - C_{12} -arylene-bis-benzamide compounds, N,N'- C_5 - C_8 -cycloalkyl-bis-benzamide compounds, N,N'-p- C_6 - C_{12} -arylene-bis- C_5 - C_8 -cycloalkylcarboxamide compounds and/or N,N'- C_5 - C_8 -cycloalkyl-bis-cyclohexanecarboxamide compounds; and/or
- amino acid derivative type diamide compounds from amidation reaction of C_5 - C_8 -alkyl-, C_5 - C_8 -cycloalkyl- or C_6 - C_{12} -aryl amino acids, C_5 - C_8 -alkyl-, C_5 - C_8 -cycloalkyl- or C_6 - C_{12} -aromatic monocarboxylic acid chlorides and C_5 - C_8 -alkyl-, C_5 - C_8 -cycloalkyl- or C_6 - C_{12} -aromatic monoamines, preferably N-phenyl-5-(N-benzoylamino)pentanamide and/or N-cyclohexyl-4-N-cyclohexylcarbonylamino benzamide, as β -nucleating agent.

[0016] Examples of N,N'-di- C_5 - C_8 -cycloalkyl-2,6-naphthalene dicarboxamide compounds are

N,N'-dicyclohexyl-2,6-naphthalene dicarboxamide and
N,N'-dicyclooctyl-2,6-naphthalene dicarboxamide.

[0017] Examples of N,N'-di- C_5 - C_8 -cycloalkyl-4,4-biphenyldicarboxamide compounds are

N,N'-dicyclohexyl-4,4-biphenyldicarboxamide and
N,N'-dicyclopentyl-4,4-biphenyldicarboxamide.

[0018] Examples of N,N'-di- C_5 - C_8 -cycloalkylterephthalamide compounds are

N,N'-dicyclohexylterephthalamide and N,N'-dicyclopentylterephthalamide.

[0019] Examples of N,N'-di- C_5 - C_8 -cycloalkyl-1,4-cyclohexanedicarboxamide compounds are

N,N'-dicyclohexyl-1,4-cyclohexanedicarboxamide and
N,N'-dicyclohexyl-1,4-cyclopentanedicarboxamide.

[0020] Examples of N,N'-di- C_6 - C_{12} -aryl- C_5 - C_8 -diamide compounds are

N,N'-bis(p-methylphenyl)-hexanediamide, N,N'-bis(4-cyclohexylphenyl)hexanediamide,
N,N'-diphenylhexanediamide, N,N'-diphenyloctanediamide and
N,N'-bis(p-ethylphenyl)hexanediamide.

[0021] Examples of N,N'- C_6 - C_{12} -arylene-bis-benzamide compounds are

N,N'-p-phenylene-bis-benzamide and N,N'-1,5-naphthalene-bis-benzamide.

[0022] Examples of N,N'-C₅-C₈-cycloalkyl-bis-benzamide compounds are

N,N'-1,4-cyclopentane-bis-benzamide and N,N'-1,4-cyclohexane-bis-benzamide.

[0023] Examples of N,N'-p-C₆-C₁₂-arylene-bis-C₅-C₈-cycloalkylcarboxamide compounds are

N,N'-1,5-naphthalene-bis-cyclohexanecarboxamide and
N,N'-1,4-phenylene-bis-cyclohexanecarboxamide.

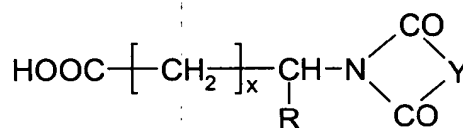
[0024] Examples of N,N'-C₅-C₈-cycloalkyl-bis-cyclohexanecarboxamide compounds are

N,N'-1,4-cyclopentane-bis-cyclohexanecarboxamide and
N,N'-1,4-cyclohexane-bis-cyclohexanecarboxamide.

[0025] According to a further preferred embodiment the β -nucleated propylene polymer contains 0.0001 to 2.0 wt% of quinacridone type compounds, especially quinacridone, dimethylquinacridone and/or dimethoxyquinacridone; quinacridonequinone type compounds, especially quinacridonequinone, a mixed crystal of 5,12-dihydro(2,3b)acridine-7,14-dione with quino(2,3b)acridine-6,7,13,14-(5H,12H)-tetrone as disclosed in EP-B 0 177 961 and/or dimethoxyquinacridonequinone; and/or dihydroquinacridone type compounds, especially dihydroquinacridone, di-methoxydihydroquinacridone and/or dibenzodihydroquinacridone, as β -nucleating agent.

[0026] According to a further preferred embodiment the β -nucleated propylene polymer contains 0.01 to 2.0 wt% of dicarboxylic acid salts of metals from group IIa of periodic system, especially pimelic acid calcium salt and/or suberic acid calcium salt; and/or mixtures of dicarboxylic acids and salts of metals from group IIa of periodic system, as β -nucleating agent.

[0027] According to a further preferred embodiment the β -nucleated propylene polymer contains 0.01 to 2.0 wt% of salts of metals from group IIa of periodic system and imido acids of the formula



wherein $x = 1$ to 4 ; $\text{R} = \text{H}$, $-\text{COOH}$, $\text{C}_1\text{-C}_{12}$ -alkyl, $\text{C}_5\text{-C}_8$ -cycloalkyl or $\text{C}_6\text{-C}_{12}$ -aryl, and $\text{Y} = \text{C}_1\text{-C}_{12}$ -alkyl, $\text{C}_5\text{-C}_8$ -cycloalkyl or $\text{C}_6\text{-C}_{12}$ -aryl - substituted bivalent $\text{C}_6\text{-C}_{12}$ -aromatic residues, especially calcium salts of phthaloylglycine, hexahydrophthaloylglycine, N-phthaloylalanine and/or N-4-methylphthaloylglycine, as β -nucleating agent.

[0028] The inventive propylene polymers with improved properties may contain usual auxiliary materials such as 0.01 to 2.5 wt% stabilizers, and/or 0.01 to 1 wt% processing aids, and/or 0.1 to 1 wt% antistatic agents and/or 0.2 to 3 wt% pigments, in each case based on the propylene polymers used.

[0029] The stabilizers, contained in the inventive propylene polymers, preferably are mixtures of 0.01 to 0.6 wt% phenolic antioxidants, 0.01 to 0.6 wt% 3-arylbenzofuranones, 0.01 to 0.6 wt% processing stabilizers based on phosphites, 0.01 to 0.6 wt% high temperature stabilizers based on disulfides and thioethers and/or 0.01 to 0.8 wt% sterically hindered amines (HALS).

[0030] A further object of the present invention is a process for producing propylene polymers with an improved property spectrum from propylene homopolymers with melt indices of 0.05 to 15 g/10 min at 230°C/2.16 kg and/or block copolymers from 90.0 to 99.9 wt% of propylene and 0.1 to 10 wt% of α -olefins with 2 or 4 to 18 carbon atoms with melt indices of 0.05 to 20 g/10 min at 230°C/2.16 kg, wherein β -nucleated propylene homopolymers have an $\text{IR}_T > 0.98$, a tensile modulus of > 1500 MPa at +23 °C and a Charpy impact strength of ≥ 3 kJ/m² at -20 °C using notched test specimens, respectively β -nucleated propylene block copolymers having an IR_T of the propylene homopolymer block of 0.98, a tensile modulus of ≥ 1100 MPa at +23 °C and a Charpy impact strength, > 6 kJ/m² at -20 °C using notched test specimens, are produced by melt mixing of propylene homopolymers having an $\text{IR}_T > 0.98$, respectively propylene block copolymers having an IR_T of the propylene homopolymer block of > 0.98 , with 0.0001 to 2.0 wt%, based on the polypropylenes used, of β -nucleating agents at temperatures from 175 to 250°C.

[0031] For the melt mixing of the propylene polymers and the β -nucleating agents, conventional compounding extruders are used.

[0032] In producing the inventive propylene polymers the β -nucleating agents may be introduced into the propylene polymers as master batch for example from 0.05 to 25 parts by weight of β -nucleating agent and 75 to 99.95 parts by weight of propylene polymer.

[0033] Preferred applications of the propylene polymers with improved properties as well as of mixtures with conventional propylene homopolymers and/or propylene copolymers are molded parts in a pipe system, such as pipes and fittings, inspection chambers, pipe ducting systems, extrusion or compression molded sheets and the like.

Examples

[0034] The following tests were made using injection molded samples prepared according to ISO 1873 Tensile modulus according to ISO 527 (cross head speed 1 mm/min) at +23°C Charpy impact strength using notched specimens, according to ISO 179/1eA

Example 1

[0035] A mixture of 90 wt% of a propylene block copolymer, obtained by combined bulk and gas phase polymerization using a Ziegler-Natta catalyst system with dicyclopentylmethoxysilane as external donor, having an ethylene content of 8.3 wt%, an IRT of the propylene homopolymer block of 0.98, and a melt index of 0.30 g/10 min at 230°C/2.16 kg, 10 wt% of a master batch comprising 99 parts by weight of a propylene block copolymer having an ethylene content of 8.3 wt%, an IRT of the propylene homopolymer block of 0.985 and a melt index of 0.30 g/10 min at 230°C/2.16 kg, and 1 part by weight of pimelic acid calcium salt and 0.1 wt% calcium stearate, 0.1 wt% tetrakis[methylene(3,5-di-*t*-butylhydroxyhydrocinnamate)]methane and 0.1 wt% tris-(2,4-di-*t*-butylphenyl)phosphite, based on the sum of the propylene polymers used, is melted in a twin screw extruder with a temperature profile of 100/145/185/210/220/200/185°C, homogenized, discharged and pelletized.

[0036] The resulting polypropylene polymer has a melt index of 0.32 g/10 min at 230°C/2.16 kg, a tensile modulus of 1290 MPa and a Charpy impact strength, notched, of 39 kJ/m² at -20°C.

Example 2

[0037] A mixture of 94 wt% of a propylene homopolymer, obtained by bulk polymerization using a Ziegler-Natta catalyst system with dicyclopentylmethoxysilane as external donor, having an IRT of 0.985 and a melt index of 1.1 g/10 min at 230°C/2.16 kg, 6 wt% of a master batch comprising 98.8 parts by weight of a propylene block copolymer having an ethylene content of 8.3 wt%, an IRT of the propylene homopolymer block of 0.985 and a melt index of 0.30 g/10 min at 230°C/2.16 kg, and 0.2 parts by weight of a mixed crystal of 5,12-dihydro(2,3b)acridine-7,14-dione with quino(2,3b)acridine-6,7,13,14-(5H,12H)-tetrone and 0.05 wt% calcium stearate, 0.1 wt% tetrakis[methylene(3,5-di-*t*-butylhydroxyhydrocinnamate)]methane and 0.1 wt% tris-(2,4-di-*t*-butylphenyl)-phosphite, based on the sum of the propylene polymers used is melted in a twin screw extruder with a temperature profile of 100/145/190/215/225/205/190°C, homogenized, discharged and pelletized.

[0038] The resulting polypropylene polymer has a melt index of 1.0 g/10 min at 230°C/2.16 kg, a tensile modulus of 1500 MPa and a Charpy impact strength, notched, of 11 kJ/m² at -20°C.

Example 3

[0039] A mixture of 75 wt% of a propylene block copolymer, obtained by combined bulk and gas phase polymerization using a Ziegler-Natta catalyst system with dicyclopentylmethoxysilane as external donor, having an ethylene content of 8.3 wt%, an IRT of the propylene homopolymer block of 0.985 and a melt index of 0.30 g/10 min at 230°C/2.16 kg, 25 wt% of a master batch comprising 99.5 parts by weight of a propylene block copolymer having an ethylene content of 8.3 wt%, an IRT of the propylene homopolymer block of 0.987 and a melt index of 0.30 g/10 min at 230°C/2.16 kg, and 0.5 parts by weight of hexahydrophthaloylglycine calcium salt and 0.1 wt% calcium stearate, 0.1 wt% tetrakis[methylene(3,5-di-*t*-butylhydroxyhydrocinnamate)]methane and 0.1 wt% tris-(2,4-di-*t*-butylphenyl)phosphite, based on the sum of the propylene polymers used, is melted in a twin screw extruder with a temperature profile of 100/145/185/210/220/200/ 185°C, homogenized, discharged and pelletized.

[0040] The resulting polypropylene polymer has a melt index of 0.32 g/10 min at 230°C/2.16 kg, a tensile modulus of 1310 MPa and a Charpy impact strength, notched, of 37 kJ/m² at -20°C.

Example 4

[0041] A mixture of

95 wt% of a propylene homopolymer, obtained by bulk phase polymerization using a Ziegler-Natta catalyst system with dicyclopentyldimethoxysilane as external donor, having an $IR\tau$ of 0.987, and a melt index of 1.1 g/10 min at 230°C/2.16 kg,

5 wt% of a master batch comprising 97.5 parts by weight of a propylene homopolymer having an $IR\tau$ of 0.987 and a melt index of 4.2 g/10 min at 230°C/2.16 kg, and 2.5 parts by weight of N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide and 0.05 wt% calcium stearate, 0.1 wt% tetrakis[methylene(3,5-di-t-butylhydroxyhydrocinnamate)]methane and 0.1 wt% tris-(2,4-di-t-butylphenyl)-phosphite, based on the sum of the propylene polymers used, is melted in a twin screw extruder with a temperature profile of 100/145/190/215/225/205/190°C, homogenized, discharged and pelletized.

[0042] The resulting polypropylene polymer has a melt index of 1.2 g/10 min at 230°C/2.16 kg, a tensile modulus of 1765 MPa and a Charpy impact strength, notched, of 5.5 kJ/m² at -20°C.

Example 5

[0043] A mixture of

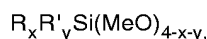
95 wt% of a propylene homopolymer, obtained by bulk phase polymerization using a Ziegler-Natta catalyst system with dicyclopentyldimethoxysilane as external donor, having an $IR\tau$ of 0.987, and a melt index of 0.3 g/10 min at 230°C/2.16 kg,

5 wt% of a master batch comprising 97.5 parts by weight of a propylene homopolymer having an $IR\tau$ of 0.987 and a melt index of 0.3 g/10 min at 230°C/2.16 kg, and 2.5 parts by weight of N,N'-dicyclohexyl-2,6-naphthalenedicarboxamide and 0.05 wt% calcium stearate, 0.1 wt% tetrakis[methylene(3,5-di-t-butylhydroxyhydrocinnamate)]methane and 0.1 wt% tris-(2,4-di-t-butylphenyl)-phosphite, based on the sum of the propylene polymers used, is melted in a twin screw extruder with a temperature profile of 100/145/190/215/225/205/190°C, homogenized, discharged and pelletized.

[0044] The resulting polypropylene polymer has a melt index of 0.3 g/10 min at 230°C/2.16 kg, a tensile modulus of 1750 MPa and a Charpy impact strength, notched, of 8.4 kJ/m² at -20°C.

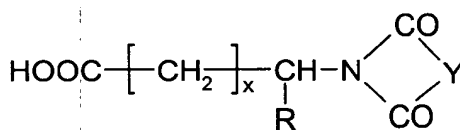
Claims

1. Propylene polymers with improved properties comprising propylene homopolymers with melt indices of 0.05 to 15 g/10 min at 230°C/2.16 kg or propylene block copolymers with 90.0 to 99.9 wt% propylene and 0.1 to 10 wt% α -olefins with 2 or 4 to 18 carbon atoms with melt indices of 0.05 to 20 g/10 min at 230 °C/2.16 kg, or mixtures thereof, wherein the propylene homopolymers or propylene block copolymers are β -nucleated propylene polymers, whereby the β -nucleated propylene homopolymers have an $IR\tau$ >0.98, a tensile modulus of ≥ 1500 MPa at +23°C and a Charpy impact strength of ≥ 3 kJ/m² at -20°C using notched test specimens, and the β -nucleated propylene block copolymers are polymers having an $IR\tau$ of the propylene homopolymer block of >0.98, a tensile modulus of ≥ 1100 MPa at +23 °C and a Charpy impact strength, of ≥ 6 kJ/m² at -20°C using notched test specimens.
2. Propylene polymers according to claim 1 comprising propylene homopolymers with melt indices of 0.1 - 8 g/10 min at 230°C/2.16 kg or propylene block copolymers with 90.0 to 99.9 wt% propylene and 0.1 to 10 wt% α -olefins with 2 or 4 to 18 carbon atoms with melt indices of 0.1 to 8 g/10 min at 230°C/2.16 kg, or mixtures thereof, wherein the propylene homopolymers or propylene block copolymers are β -nucleated propylene polymers, whereby the β -nucleated propylene homopolymers have an $IR\tau$ >0.98, a tensile modulus of ≥ 1600 MPa at +23°C and a Charpy impact strength of 4 to 10 kJ/m² at -20°C using notched test specimens, and the β -nucleated propylene block copolymers are polymers having an $IR\tau$ of the propylene homopolymer block of >0.98, a tensile modulus of ≥ 1300 MPa at +23 °C and a Charpy impact strength, of ≥ 9 kJ/m² at -20°C using notched test specimens.
3. Propylene polymers according to one of the claims 1 or 2, wherein the β -nucleated propylene homopolymers with an $IR\tau$ >0.98 or propylene block copolymers with an $IR\tau$ >0.98 in the homopolymer block are propylene polymers obtained by polymerization with a Ziegler-Natta catalyst system comprising titanium-containing solid components, an organoalumina, magnesium or titanium compound as cocatalyst and an external donor according to the formula



wherein R and R' are identical or different and are branched or cyclic aliphatic or aromatic hydrocarbon residues, and y and x independently from each other are 0 or 1, provided that x + y are 1 or 2.

4. Propylene polymers according to claim 3, **characterized in that** the external donor is dicyclopentyl dimethoxysilane.
5. Propylene polymers according to one of the claims 1 to 4, wherein the β -nucleated propylene polymer contains 0.01 to 2.0 wt%, in each case based on the propylene polymers used, of dicarboxylic acid derivative type diamide compounds from C₅-C₈-cycloalkyl monoamines or C₆-C₁₂-aromatic monoamines and C₅-C₈-aliphatic, C₅-C₈-cycloaliphatic or C₆-C₁₂-aromatic dicarboxylic acids, preferably N,N'-di-C₅-C₈-cycloalkyl-2,6-naphthalene dicarboxamide compounds, N,N'-di-C₅-C₈-cycloalkyl-4,4-biphenyldicarboxamide compounds, N,N'-di-C₅-C₈-cycloalkylterephthalamide compounds, N,N'-di-C₅-C₈-cycloalkyl-1,4-cyclohexanedicarboxamide compounds and/or N,N'-di-C₆-C₁₂-aryl-C₅-C₈-diamide compounds; and/or diamine derivative type diamide compounds from C₅-C₈-cycloalkyl monocarboxylic acids or C₆-C₁₂-aromatic monocarboxylic acids and C₅-C₈-cycloaliphatic or C₆-C₁₂-aromatic diamines, preferably N,N'-C₆-C₁₂-arylene-bis-benzamide compounds, N,N'-C₅-C₈-cycloalkyl-bis-benzamide compounds, N,N'-p-C₆-C₁₂-arylene-bis-C₅-C₈-cycloalkylcarboxamide compounds and/or N,N'-C₅-C₈-cycloalkyl-bis-cyclohexanecarboxamide compounds; and/or amino acid derivative type diamide compounds from amidation reaction of C₅-C₈-alkyl-, C₅-C₈-cycloalkyl- or C₆-C₁₂-aryl amino acids, C₅-C₈-alkyl-, C₅-C₈-cycloalkyl- or C₆-C₁₂-aromatic monocarboxylic acid chlorides and C₅-C₈-alkyl-, C₅-C₈-cycloalkyl- or C₆-C₁₂-aromatic monoamines, preferably N-phenyl-5-(N-benzoylamino)pentaneamide and/or N-cyclohexyl-4-N-cyclohexylcarbonylamino benzamide, as β -nucleating agent.
6. Propylene polymers according to one of the claims 1 to 4, **characterized in that** the β -nucleated propylene polymer contains 0.0001 to 2.0 wt% quinacridone type compounds, preferably quinacridone, dimethylquinacridone and/or dimethoxyquinacridone; quinacridonequinone type compounds, preferably quinacridonequinone, a mixed crystal of 5,12-dihydro(2,3b)acridine-7,14-dione with quino(2,3b)acridine-6,7,13,14-(5H,12H)-tetrone and/or dimethoxyquinacridonequinone; and/or dihydroquinacridone type compounds, preferably dihydroquinacridone, dimethoxydihydroquinacridone and/or dibenzodihydroquinacridone, as β -nucleating agent.
7. Propylene polymers according to one of the claims 1 to 4, **characterized in that** the β -nucleated propylene polymer contains 0.01 to 2.0 wt% dicarboxylic acid salts of metals from group IIa of periodic system, preferably pimelic acid calcium salt and/or suberic acid calcium salt; and/or mixtures of dicarboxylic acids and metals from group IIa of periodic system, as β -nucleating agent.
8. Propylene polymers according to one of the claims 1 to 4, **characterized in that** the β -nucleated propylene polymer contains 0.01 to 2.0 wt% of salts of metals from group IIa of periodic system and imido acids of the formula



wherein x = 1 to 4; R = H, -COOH, C₁-C₁₂-alkyl, C₅-C₈-cycloalkyl or C₆-C₁₂-aryl, and Y = C₁-C₁₂-alkyl, C₅-C₈-cycloalkyl or C₆-C₁₂-aryl - substituted bivalent C₆-C₁₂-aromatic residues, preferably calcium salts of phthaloylglycine, hexahydrophthaloylglycine, N-phthaloylalanine and/ or N-4-methylphthaloylglycine, as β -nucleating agent.

9. A process for producing propylene polymers with improved properties from propylene homopolymers with melt indices of 0.05 to 15 g/10 min at 230°C/2.16 kg and/or block copolymers from 90.0 to 99.9 wt% of propylene and 0.1 to 10 wt% of α -olefins with 2 or 4 to 18 carbon atoms with melt indices of 0.05 to 20 g/10 min at 230°C/2.16 kg, wherein β -nucleated propylene homopolymers have an IR τ > 0.98, a tensile modulus \geq 1500 MPa and a Charpy impact strength of \geq 3 kJ/m² at -20°C, using notched test specimens, respectively β -nucleated propylene block copolymers having an IR τ of the propylene homopolymer block of > 0.98, a tensile modulus \geq 1100 MPa and a Charpy impact strength > 6 kJ/m², at -20°C, using notched test specimens, are produced by melt mixing of propylene homopolymers having an IR τ > 0.98, respectively propylene block copolymers having an IR τ of the propylene homopolymer block of > 0.98, with 0.0001 to 2.0 wt%, based on the polypropylenes used, of β -nucleating agents

at temperatures from 175 to 250°C.

10. Use of the propylene polymers with an improved properties according to one of the claims 1 to 8 as well as of mixtures with conventional propylene homopolymers and/or propylene copolymers for producing molded parts in a pipe system, such as pipes and fittings, inspection chambers, pipe ducting systems, extrusion or compression molded sheets and the like.

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European Patent
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EUROPEAN SEARCH REPORT

Application Number
EP 01 11 2367

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
X	WO 99 24479 A (BOREALIS AS ;JAEAESKELAEINEN PIRJO (FI); MALM BO (FI); HUOVINEN PA) 20 May 1999 (1999-05-20) * claims; examples; tables *	1-4,9,10	C08F110/06 C08F297/08 C08K5/00
D,X	EP 0 277 514 A (CHISSO CORP) 10 August 1988 (1988-08-10) * column 2, line 8 - line 31; claims; examples 2-5,10,11; tables *	1-4,9	
D,X	EP 0 177 961 A (DANUBIA PETROCHEMIE) 16 April 1986 (1986-04-16) * claims; examples; table 3 *	1-4,6,9	
X	EP 0 714 923 A (SHOWA DENKO KK) 5 June 1996 (1996-06-05) * page 7, line 48 - page 8, line 5; claims; examples; tables *	1-4,9,10	
A	WO 97 13790 A (BOREALIS AS ;ANDTSJOE HENRIK (FI); PENTTI ISMO (FI); HARLIN ALI (F) 17 April 1997 (1997-04-17) * page 11, line 16 - line 20; claims; examples 6,8 *	1-3,9,10	TECHNICAL FIELDS SEARCHED (Int.Cl.7) C08F C08L C08K
D,A	EP 0 790 262 A (CHISSO CORP) 20 August 1997 (1997-08-20) * page 8, line 50 - page 18, line 56; claims *	1-9	
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 23 October 2001	Examiner Kaumann, E
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

EPC FORM 1503 03 02 (P04C01)

**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 01 11 2367

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

23-10-2001

Patent document cited in search report		Publication date		Patent family member(s)	Publication date
WO 9924479	A	20-05-1999	AU	1035799 A	31-05-1999
			BR	9814854 A	03-10-2000
			CN	1285851 T	28-02-2001
			EP	1028985 A1	23-08-2000
			WO	9924479 A1	20-05-1999
EP 0277514	A	10-08-1988	JP	7005668 B	25-01-1995
			JP	63191809 A	09-08-1988
			CA	1300312 A1	05-05-1992
			CZ	8800680 A3	16-08-1995
			DE	3885912 D1	13-01-1994
			DE	3885912 T2	28-04-1994
			EP	0277514 A2	10-08-1988
			HK	60295 A	28-04-1995
			KR	9601217 B1	24-01-1996
			SG	9590591 A2	01-09-1995
			SK	68088 A3	10-12-1997
			US	4981938 A	01-01-1991
EP 0177961	A	16-04-1986	AT	381110 A	25-08-1986
			AT	322984 A	15-01-1986
			CS	8507292 A3	18-11-1992
			DE	3443599 A1	17-04-1986
			DE	3561399 D1	18-02-1988
			EP	0177961 A2	16-04-1986
			HU	40152 A2	28-11-1986
			SK	277920 B6	09-08-1995
EP 0714923	A	05-06-1996	AU	705455 B2	20-05-1999
			AU	3790395 A	30-05-1996
			CA	2162946 A1	22-05-1996
			CN	1128767 A	14-08-1996
			DE	69517771 D1	10-08-2000
			DE	69517771 T2	23-11-2000
			EP	0714923 A1	05-06-1996
			JP	3002119 B2	24-01-2000
			JP	8198922 A	06-08-1996
			US	5703172 A	30-12-1997
			US	5804665 A	08-09-1998
			JP	3034452 B2	17-04-2000
			JP	8198923 A	06-08-1996
WO 9713790	A	17-04-1997	FI	954814 A	11-04-1997
			AT	188711 T	15-01-2000
			AU	704614 B2	29-04-1999
			AU	7218296 A	30-04-1997

EPO FORM P0453

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 01 11 2367

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

23-10-2001

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9713790 A		BR 9610966 A	02-03-1999
		CN 1202177 A	16-12-1998
		DE 69606188 D1	17-02-2000
		DE 69606188 T2	08-06-2000
		EP 0854887 A1	29-07-1998
		ES 2142615 T3	16-04-2000
		WO 9713790 A1	17-04-1997
		US 6084041 A	04-07-2000
		ZA 9608558 A	20-05-1997
EP 0790262 A	20-08-1997	AU 717990 B2	06-04-2000
		AU 6838296 A	19-03-1997
		DE 69608240 D1	15-06-2000
		DE 69608240 T2	23-11-2000
		EP 0790262 A1	20-08-1997
		US 6005034 A	21-12-1999
		CA 2203876 A1	06-03-1997
		WO 9708218 A1	06-03-1997
		TW 416962 B	01-01-2001

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ASSIGNEE-INFORMATION:

NAME	COUNTRY
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C08K005/00

ABSTRACT:

CHG DATE=20030114 STATUS=O> The invention relates to novel propylene polymers with improved properties especially with improved stiffness and impact strength comprising propylene homopolymers or propylene block copolymers with 90.0 to 99.9 wt % propylene and 0.1 to 10 wt% alpha -olefins with 2 or 4 to 18 carbon atoms, or mixtures thereof,

wherein the propylene homopolymers or propylene block copolymers are beta -nucleated propylene polymers, whereby the beta -nucleated propylene homopolymers have an IR tau >0.98, a tensile modulus >1500 MPa at +23 DEG C and a Charpy impact strength, ≥ 3 kJ/m² at -20 DEG C using notched test specimens, and the beta -nucleated propylene block copolymers are polymers having an IR tau of the propylene homopolymer block of >0.98, a tensile modulus ≥ 1100 MPa at +23 DEG C and a Charpy impact strength of >6kJ/m², at -20 DEG C using notched test specimens. The propylene polymers with an improved property spectrum are suitable for producing molded parts in a pipe system, such as pipes and fittings, inspection chambers, pipe ducting systems, extrusion or compression molded sheets and the like.